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**FLUX PINNING ENHANCEMENT IN  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$  FILMS  
WITH  $\text{BaSnO}_3$  NANOPARTICLES (POSTPRINT)**

**Chakrapani V. Varanasi, P.N. Barnes, J. Burke, L. Brunke, I. Maartense, T.J. Haugan,  
E.A. Stinzianni, K.A. Dunn, and P. Haldar**

**Power Generation Branch  
Power Division**

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PROPULSION DIRECTORATE  
WRIGHT-PATTERSON AIR FORCE BASE, OH 45433-7251  
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UNITED STATES AIR FORCE**



## RAPID COMMUNICATION

# Flux pinning enhancement in $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ films with $\text{BaSnO}_3$ nanoparticles

Chakrapani V Varanasi<sup>1</sup>, P N Barnes<sup>2</sup>, J Burke<sup>1</sup>, L Brunke<sup>1</sup>,  
I Maartense<sup>1</sup>, T J Haugan<sup>2</sup>, E A Stinzianni<sup>3</sup>, K A Dunn<sup>3</sup> and  
P Haldar<sup>3</sup>

<sup>1</sup> University of Dayton Research Institute (UDRI), Dayton, OH, USA

<sup>2</sup> Air Force Research Laboratory (AFRL), Wright-Patterson AFB, OH, USA

<sup>3</sup> College of Nanoscale Science and Engineering, University at Albany, State University of New York, USA

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## Abstract

Nanoparticles of  $\text{BaSnO}_3$  were incorporated into  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$  (YBCO) films on  $\text{LaAlO}_3$  substrates for magnetic flux pinning enhancements. More than an order of magnitude improvement in the high field magnetization  $J_c$  at 6 T at 77 K was observed as compared to regular YBCO films. The irreversibility field ( $H_{irr}$ ) was increased to 8.5 T at 77 K and to 13.4 T at 65 K. The in-field transport current measurements confirmed an order of magnitude improvement in high fields. The angular dependence of the  $J_c$  data at 1 T showed that  $J_c H \parallel c$  is 1.3 times higher than  $J_c H \parallel ab$  indicating the presence of  $c$ -axis correlated defects. Transmission electron microscopy studies revealed the presence of a large density of uniformly distributed  $\sim 10$  nm sized  $\text{BaSnO}_3$  precipitates and strain fields around them. A dual sector pulsed laser deposition target is used to produce the films, thus eliminating reactions between  $\text{BaSnO}_3$  and YBCO during the target preparation stage, but may allow the  $\text{BaSnO}_3$  to react locally and create defects that act as pinning centres.

(Some figures in this article are in colour only in the electronic version)

## 1. Introduction

$\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$  (YBCO) coated conductors are processed by depositing YBCO coatings on buffered highly textured metallic substrates. These metallic substrates can be either textured [1] in order to transfer the texture to the buffer layers or polished polycrystalline substrates on which buffer layers were grown with texture using ion beam assisted deposition (IBAD) [2]. Significant improvements have been achieved in the processing of textured YBCO coated conductors in long lengths which can carry large currents [3]. However, additional improvement in the critical current density ( $J_c$ ) of YBCO is needed as a means to make a higher in-field engineering current density ( $J_E$ ) conductor necessary for its

application in devices [4]. Increasing the  $J_c$  at higher fields can be accomplished by introducing artificial magnetic flux pinning centres into YBCO coatings. The pinning centres, in order to be effective, need to be nanoparticles, less than 15 vol% of the superconductor, and be randomly distributed to provide isotropic 3D pinning. Various materials such as  $\text{Y}_2\text{O}_3$ ,  $\text{Y}_2\text{BaCuO}_5$ ,  $\text{BaZrO}_3$ ,  $\text{BaIrO}_3$ ,  $\text{Nd}_2\text{O}_3$  etc [5–12] in pulsed laser ablated (PLD) YBCO films have been recently investigated for creating nanoparticles for flux pinning. The pinning effect of each of these materials and processes provides effective pinning at various magnetic field levels.

One way to introduce second phase particles in PLD YBCO films is by the use of a doped YBCO target. YBCO powder is blended with a desired amount of second phase

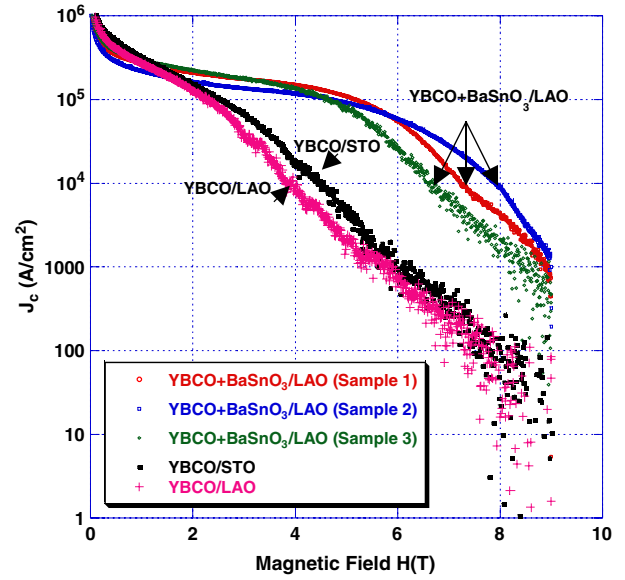
materials and sintered together to form a composite target of superconductor and pinning material. However, reactions with the pinning material and YBCO during the target preparation are possible in this approach if the pinning material is not chemically compatible with the YBCO. In addition, targets with dissimilar melting point compositions (e.g. low melting point metal + YBCO) will be hard to make. In this work, a special PLD dual phase sector target was used to introduce  $\text{BaSnO}_3$  nanoparticles into YBCO films and the properties were investigated. Such a method was demonstrated to create nanometre sized  $\text{Y}_2\text{BaCuO}_5$  particles in YBCO films successfully in a previous study [7].

$\text{BaSnO}_3$  with a cubic perovskite crystal type structure ( $a = 4.12 \text{ \AA}$ ) has been investigated as a buffer layer on  $\text{MgO}$  substrates for growing smoother YBCO films for microwave applications [13]. Although  $\text{BaSnO}_3$  has been studied for flux pinning enhancements in melt processed YBCO [14, 15], here we provide an initial demonstration of effectively incorporating  $\text{BaSnO}_3$  nanoparticles as pinning material in YBCO films. The processing temperatures and method of  $\text{BaSnO}_3$  nanoparticle introduction into YBCO thin films are quite different from melt processing methods. Due to a higher lattice mismatch and probable slight reactivity with the YBCO matrix (possible Sn diffusion into YBCO etc), the defects surrounding the nanoparticles created by the  $\text{BaSnO}_3$  particles may well be potentially different from the other materials have been used earlier and may provide an enhanced flux pinning effectiveness at high fields.

## 2. Experimental details

A specially made YBCO pulsed laser ablation target with a  $\text{BaSnO}_3$  second phase pie wedge/sector was used to deposit the YBCO +  $\text{BaSnO}_3$  coatings. The details of the dual phase sector PLD deposition are discussed elsewhere [7]. Briefly, a target that consists of  $\text{BaSnO}_3$  and YBCO sectors, as in a pie diagram, is made to rotate during the deposition. This allows the ablation of  $\text{BaSnO}_3$  periodically, allowing the introduction of nanoparticles in a growing YBCO film. The angle of the  $\text{BaSnO}_3$  sector used in the present work is  $30^\circ$ . Per target rotation, the  $\text{BaSnO}_3$  sector gets ablated once in approximately every 12 laser pulses. A Lambda Physik KrF excimer laser (wavelength  $\lambda = 248 \text{ nm}$ ) was used to deposit films in a Neocera PLD chamber. The deposition was carried out using  $2 \text{ J cm}^{-2}$  laser energy at a 4 Hz repetition rate with a substrate temperature maintained at  $780^\circ\text{C}$ . The target to heater distance was 6 cm. The target was rotated at a speed of 15–20 rpm to obtain the composite YBCO +  $\text{BaSnO}_3$  nanoparticle film. Films were deposited on (100) lanthanum aluminate single crystal substrates ( $\text{LaAlO}_3$ ) to investigate the flux pinning improvements.

The critical transition temperature ( $T_c$ ) of the films was measured by an ac susceptibility method. The film microstructure was studied by using a SIRION high resolution scanning electron microscope (SEM) and a high resolution transmission electron microscope (TEM; JEOL, model 2010-F) operated at 200 kV and equipped with an energy dispersive x-ray spectrometer (EDS). Cross-sectional and plan view specimens were prepared using a hybrid method combining focused ion beam lift-out and conventional  $\text{Ar}^+$  ion milling for



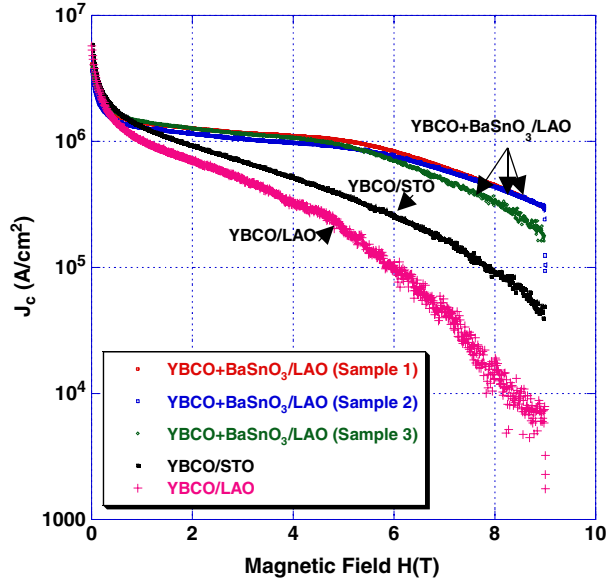
**Figure 1.** Magnetization  $J_c$  at 77 K for YBCO +  $\text{BaSnO}_3$  samples on  $\text{LaAlO}_3$  substrates compared with YBCO control samples on  $\text{LaAlO}_3$  and  $\text{SrTiO}_3$  substrates.

final thinning. The magnetization  $J_c$  was measured by using a Quantum Design PPMS vibrating sample magnetometer (VSM) at 77 and 65 K. The thickness of the films was measured by using a profilometer and verified with a cross-sectional SEM and used for  $J_c$  calculations. Transport current measurements were taken on a  $\sim 0.3 \text{ }\mu\text{m}$  thick, 3 mm long, 1 mm wide bridge sample and in-field measurements performed to 12 T. The angular dependence of  $J_c$  at 1 T was also measured.

## 3. Results and discussion

The  $T_c$  of the YBCO +  $\text{BaSnO}_3$  films was found to be slightly reduced (between 86 and 89 K) as compared to regular YBCO films which are routinely made with  $T_c$ s higher than 90 K on  $\text{LaAlO}_3$  substrates. The lowering of  $T_c$  in the presence of the nanoparticles is consistent with other studies done with  $\text{BaZrO}_3$  nanoparticles in PLD YBCO films. The reason for the depressed  $T_c$  may be possible Sn diffusion into the YBCO which might result in Sn substitutions in the copper sites that can locally depress  $T_c$ , as observed in the  $\text{YBa}_2\text{Cu}_{3-x}\text{Sn}_x\text{O}_{7-d}$  system [16]. The films used in the present study had a  $T_c$  of 88.5 K.

Figures 1 and 2 show the magnetization  $J_c$  data at 77 and 65 K, respectively, which were collected using a VSM. Three different YBCO +  $\text{BaSnO}_3$  samples of different thickness (240, 317, 359 nm) were compared with two standard  $\sim 300 \text{ nm}$  thick YBCO films on a (100)  $\text{SrTiO}_3$  single crystal substrate and a (100) single crystal  $\text{LaAlO}_3$  substrate. A significant increase in the magnetization  $J_c$  in YBCO +  $\text{BaSnO}_3$  films was observed at both 77 and 65 K, especially at magnetic fields  $> 2 \text{ T}$ , compared to control samples. Compared to regular YBCO, more than an order of magnitude increase in  $J_c$  at 6 T at 77 K was achieved. The increase in  $J_c$  at high fields observed with  $\text{BaSnO}_3$  particles was found to be higher than that observed with YBCO +  $\text{Y}_2\text{BaCuO}_5$  samples made by

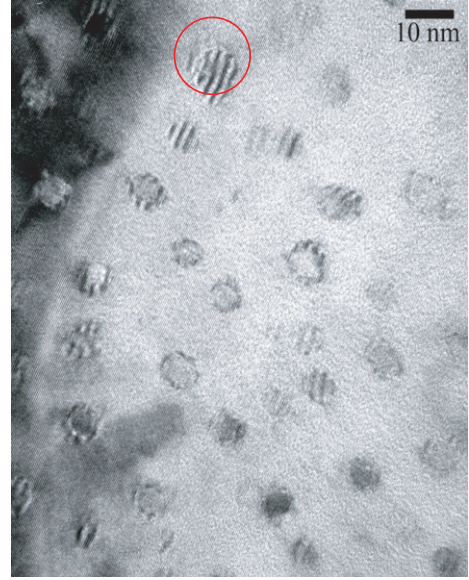


**Figure 2.** Magnetization  $J_c$  at 65 K for YBCO + BaSnO<sub>3</sub> samples on LaAlO<sub>3</sub> substrates compared with YBCO control samples on LaAlO<sub>3</sub> and SrTiO<sub>3</sub> substrates.

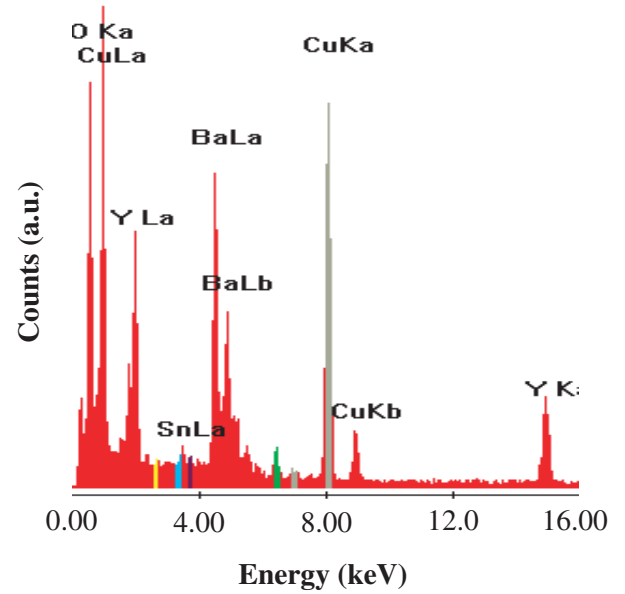
the same method [7] indicating that the defects created by the BaSnO<sub>3</sub> contribute more effectively to the flux pinning enhancement at high fields. The decrease in  $J_c$  at low fields is due to the poor crystal quality (strained YBCO crystal planes) caused by the nanoparticles or simply that optimization of the pinning density is possible. Also the incorporation of non-superconducting BaSnO<sub>3</sub> phase (estimated to be 10–15%) should lower the  $J_c$  by about 10–15% as a first approximation (due to dilution of superconducting phase) at low field. A similar decrease in the self-field  $J_c$  was also observed in SmBCO films with low  $T_c$  nanoparticles although higher  $J_c$ s were observed at high fields [17].

The  $H_{irr}$  was significantly increased for the YBCO + BaSnO<sub>3</sub> films at both 65 and 77 K as compared to YBCO. The value of  $H_{irr}$  for YBCO + BaSnO<sub>3</sub> films was found to be 8.51 T at 77 K as compared to 6–7 T commonly found for YBCO in the literature [18, 19]. The  $H_{irr}$  for the YBCO + BaSnO<sub>3</sub> films was found to be 13.4 T at 65 K. Although high irreversibility fields in SmBCO are possible [17], a high  $H_{irr}$  value of 8.5 T at 77 K for doped YBCO is determined in this study. The  $H_{irr}$  values were determined by extrapolating the linear section in the volume flux pinning force ( $F_p$ ) versus  $H$  plots. For some of the samples, evidence of twin peaks in the flux pinning force ( $F_p$ ) plots was also observed and these are believed to be due to low  $T_c$  regions in a high  $T_c$  matrix of YBCO.

SEM observations of the surface show the presence of particles typical of PLD films as well as a high density of uniformly distributed nanoparticles  $\sim 10$  nm in size. The number density of these particles was found to be  $\sim 3 \times 10^{15} \text{ m}^{-2}$ . Figure 3 displays the TEM images of the YBCO + BaSnO<sub>3</sub> samples where the nanoparticles are the striped features  $\sim 10$  nm in size spaced apart by about 10 nm. The distinct moiré fringe (stripe) contrast arises from the superposition of two disparate lattices: in this case, the nanoparticles and the YBCO matrix. All the secondary phase



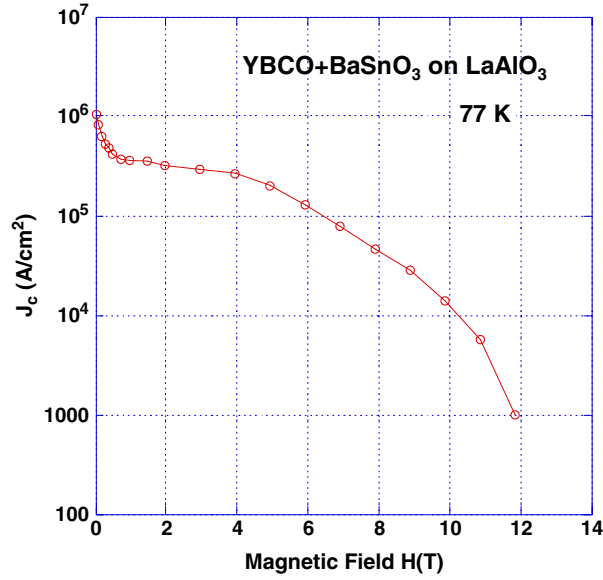
**Figure 3.** Plan view transmission electron micrograph of the YBCO + BaSnO<sub>3</sub> samples showing the presence of 10 nm particles.



**Figure 4.** X-ray energy dispersive spectra taken from one of the particles (the circled one in figure 3) showing the presence of Sn in particles.

precipitates were found to be Sn rich by EDS analysis, shown in figure 4. The cross-sectional high resolution TEM images (not shown) suggest the presence of disc-shaped defects surrounded by significant strain contrast. The strain in the lattice could be from either groups of Sn rich atoms, ultrafine BaSnO<sub>3</sub> particles or some other defects. In addition to the diffraction reflections from YBCO, additional diffraction spots were observed which may correspond to barium tin oxides but could not be uniquely attributed to the stoichiometric BaSnO<sub>3</sub> phase. The presence of these particles and the strain fields surrounding these particles due to the lattice mismatch ( $\sim 7.7\%$ ) between BaSnO<sub>3</sub> and YBCO are thought to contribute to the improvements observed in  $J_c$ .





**Figure 5.** Transport critical current density measurements on one of the YBCO + BaSnO<sub>3</sub> samples in an applied magnetic field ( $H \parallel c$ ).

In the low field regime, where  $J_c \propto H^{-\alpha}$ , the value of  $\alpha$  for typical YBCO control samples was around 0.5, similar to the value found by other groups [20]. This value indicates the presence of a dilute distribution of defects. However, with YBCO + BaSnO<sub>3</sub> films, an  $\alpha$  of 0.2–0.3 at 77 K was measured. A value of 0.3 for  $\alpha$  is consistent with the value of  $\alpha$  that was observed in other strongly pinning systems with Y<sub>2</sub>O<sub>3</sub>, BaZrO<sub>3</sub> particles [20, 8].

The transport current measurements in the applied magnetic field are shown in figure 5. It can be seen that  $J_c$  does not decrease rapidly with applied field as observed in the magnetization  $J_c$  measurements. While the self-field  $J_c$  was a little low ( $10^6$  A cm<sup>-2</sup>), a high  $J_c$  of  $1.1 \times 10^5$  A cm<sup>-2</sup> at 6 T and a  $J_c$  of  $1.1 \times 10^4$  A cm<sup>-2</sup> at 10 T were observed. The angular dependence of  $J_c$  measurements indicated that  $J_c H \parallel c$  ( $3.7 \times 10^5$  A cm<sup>-2</sup>) is higher than  $J_c (H \parallel ab)$  ( $2.8 \times 10^5$  A cm<sup>-2</sup>) by 1.3 times at 1 T and 77 K, implying that  $c$ -axis correlated defects may be present in the sample. The lattice mismatch between BaSnO<sub>3</sub> and YBCO is large enough to generate dislocations in thin film samples; however the particulate size in these samples is small enough that the strain may be accommodated elastically rather than plastically. Such an arrangement may still lead to a vertical stacking effect, where nanoparticles in the upper regions of the film nucleate above existing nanoparticles where there is a minimum in the local strain field.

The  $c$ -axis correlated defects created by the BaSnO<sub>3</sub> particles could help reduce the anisotropy of the critical current density. The technique of using a dual phase sectorized PLD target gives the flexibility of introducing second phase particulates in an uninterrupted, more random fashion. By selecting a proper laser scanning sequence, the desired amount of BaSnO<sub>3</sub> nanoparticles can be introduced into the growing YBCO film. Since the BaSnO<sub>3</sub> and YBCO are ablated with the same laser power and frequency, the particle growth of BaSnO<sub>3</sub> is restricted to nanoparticle size in the YBCO film. Also

since a separate sector is used instead of pre-mixed targets, any possible reactions between BaSnO<sub>3</sub> and YBCO during the target preparation stage were eliminated, but the BaSnO<sub>3</sub> is allowed to react locally (Sn may diffuse into YBCO to substitute for Cu) and create defects during growth in a scalable and continuous manner.

#### 4. Conclusions

BaSnO<sub>3</sub> nanoparticles were incorporated during the growth of YBCO films through ablation of a sintered BaSnO<sub>3</sub> target sector inserted within an YBCO target in a pulsed laser ablation chamber. YBCO + BaSnO<sub>3</sub> composite films have been shown to have  $J_c$ s more than an order of magnitude higher at high magnetic fields compared to regular YBCO. The presence of high number density 10 nm size nanoparticles and associated strain fields around them are believed to be responsible for the observed enhancements. These are initial results and further optimization in terms of content and the angle of BaSnO<sub>3</sub> sector are under way.

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